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(71) Applicant:
Linde AG

(72) Inventors:
Dr. Reiner Götz et al.

(54) Title: **Reactor**

[(57) Abstract: The reactor in accordance with the invention permits the achievement of adequate cooling preferably of all pressure-loaded metal parts. In addition, a safe, gas-tight passage from the area of a reactor pipe designed as a membrane pipe to the metal components of the reactor pipe is guaranteed. In addition, damaged reactor pipes can be replaced simply and rapidly. Furthermore, the reactor in accordance with the invention is designed as conventionally as possible, so that the fraction of ceramic components can be kept small. In addition, improved reaction control is made possible, since the introduction of the oxygen to the hydrocarbon or hydrocabeon mixture can take place over the pipe length, i.e., along the reaction pathway.

Specification

[0001] The invention pertains to a reactor that is especially suitable for the production of syntheses gas by partial oxidation.

[0002] For producing syntheses gas, a gas-tight, oxygen ion- and electron-conductive ceramic membrane is supplied on one side with a hot, oxygen-containing gas mixture. On the other side of the membrane (permeate side), the emerging oxygen is reacted with a supplied hydrocarbon or hydrocarbon mixture to form a syntheses gas.

[0003] The oxygen ion transport through such ceramic membranes, however, takes place in the desired direction only if the oxygen partial pressure on the retentate side is larger than on the permeate side.

[0004] On the permeate side of the membrane, as a result of the chemical reaction with the hydrocarbon or hydrocarbons, the partial pressure of the oxygen is very low, so that the oxygen-containing gas mixture supplied to the retentate side must only be compressed to a relatively low pressure. Usually the pressure of the syntheses gas produced is actually larger than that of the oxygen-containing gas mixture.

[0005] The optimal working or effect range of available ceramic membranes lies at temperatures between 700° and 1,110°C.

[0006] A larger number of reactor designs are known—for example reference may be made to EP-A 0,875,285—that can serve for production of syntheses gas with a ceramic membrane. However, this advantage is of the known reactor designs include the fact that they are on one hand relatively expensive to build, and on the other hand the cooling of the pressure-loaded metallic constituents required because of the high temperatures of the gas streams cannot always be guaranteed. In addition, replacement of damaged parts in the known designs is possible only with a comparatively high expense.

[0007] The goal of the present invention is to supply a reactor for production of syntheses gas that avoids the known drawbacks.

[0008] To achieve this goal, in accordance with a first alternative, a reactor is suggested with

-a reactor jacket,

-two covers closing the two ends of the reactor jacket

-wherein each cover has at least one opening

-a first pipe plate arranged in the upper area of the reactor and a second pipe plate arranged in the center area of the reactor

-an inner container that is arranged beneath the second pipe plate and defines a gas space

-wherein the inner container may have openings in its base

-at least one partition arranged essentially at a right angle to the reactor jacket, having openings, which subdivides the area between the first pipe plate and the second pipe plate into a gas space arranged above the partition and a gas space arranged below the partition

-in each case at least two openings corresponding to the gas space arranged below the partition and to the gas space defined by the inner container, wherein in the case of the gas space arranged below the partition, these are preferably arranged in the reactor jacket, and in the case of the gas space defined by the inner container, preferably an opening is arranged in the reactor jacket and openings in the base of the inner container

-at least one opening corresponding to the gas space arranged the above the partition

-several emergence pipes placed in the first pipe plate

-several reactor pipes placed in the second pipe plate

-wherein the removal pipes extend through the openings into the reactor pipes

-wherein the reactor pipes are at least partially designed as membrane pipes, and

-wherein the region of the reactor pipes designed as membrane pipes is arranged in the gas space defined by the inner container.

[0009] Furthermore to solve the above referenced task—in accordance with a second alternative—a reactor is suggested with

[0010] A reactor jacket,

-two covers closing off the two ends of the reactor jacket,

-wherein each jacket has at least one opening

-a first pipe plate arranged in the upper area of the reactor and a second pipe plate arranged in a middle area of the reactor

-an upper internal container and a lower internal container, each of which are arranged beneath the pipe plate and define a first and a second gas space

-wherein at least the upper inner container has openings in its plate

-at least one partition arranged essentially at right angles to the reactor jacket beneath the inner container, having openings, and subdividing the area between the upper inner container and the second pipe plate into a third and a fourth gas space

-in each case at least two openings corresponding to the first, second, and fourth gas space, wherein these are arranged preferably in the reactor jacket in the case of the first and the fourth gas space, and in the case of the second gas space, preferably one opening is arranged in the reactor jacket and one opening in the plate of the lower inner container

-at least one opening corresponding to the third gas space

-several removal pipes arranged in the first pipe plate

-several reactor pipes arranged in the second pipe plate

-wherein the removal pipes extend through the opening into the reactor pipes

-wherein the reactor pipes are at least partially designed as membrane pipes, and

-wherein the area of the reactor pipes designed as a membrane pipe is arranged in the second gas space.

[0011] Additional advantageous designs of the reactors in accordance with the invention are objects of the subclaims.

[0012] The reactor in accordance with the second alternative of the invention and further embodiments thereof will be explained in greater detail on the basis of the exemplified embodiment shown in the figure.

[0013] The figure shows a lateral sectional representation through one possible embodiment of the reactor in accordance with the invention. Such reactors are generally cylindrically symmetric design. They can be arranged either upright—as shown in the figure—or in any other alignment, for example horizontal. In the following the upright arrangement shown in the figure will be described.

[0014] They consist of the actual reactor jacket 1 and 1′ as well as 2 covers 2 and 5 that close off the two ends of the reactor jacket 1 and 1′. Each cover 2 and 5 has at least one opening 3 and 6. In practice the lower cover 5—contrary to the representation in the figure—is designed only as a plate. The subdivision of the reactor jacket into the areas 1 and 1′ distinguishes between a region 1 in which the reaction of the oxygen with the hydrocarbon or hydrocarbon gas mixture to form a syntheses gas takes place, whereas in the region 1′--which is also designated as the container channel—the cooling of the hot gas mixture depleted in oxygen takes place.

[0015] According to an advantageous embodiment of the reactor in accordance with the invention, on the inside of the reactor jacket 1,1′ and/or on the inside of one or both covers 2 and 5, a thermally insulating layer may be arranged.

[0016] A thermal protection insulation of this type serves to keep the temperature of the reactor jacket that is exposed to a heavy pressure load on a relatively low level.

[0017] In the interior of the reactor in its upper region a first pipe plate 8 is arranged, and in its center region a second pipe plate 30. Beneath this pipe plate an upper inner container 9 and a lower inner container 34 are arranged and define a first gas space 16 and a second gas space 38. The upper inner container 9 has openings 11 in its space, the purpose of which will be explained in the following.

[0018] Furthermore in an interior of the reactor a partition 19 arranged at right angles to its side beneath the inner container 9, containing openings 20; this subdivides the region between the upper inner container 9 and the second pipe plate 30 into a third gas space 21 and a fourth gas space 24.

[0019] To the first, the second, and the fourth gas space 16, 38, and 24, in each case at least two openings, through which the medium can enter and exit the reactor, are arranged. In the case of the first and the fourth gas space 16 and 24, respectively, the openings 12 and 13 and 25 and 26—as shown in the figure—are preferably arranged in the reactor jacket 1′, whereas in the case of the second gas space 38 preferably an opening 36 is arranged in the reactor jacket 1 and an opening 37 in the base of the lower inner container 34. The opening 13 mentioned also has assigned to it a guide panel 14 that defines an offtake space or gap 15. The third gas chamber 21 has only a single opening 22 arranged in the reactor jacket 1′ assigned to it.

[0020] In the first pipe plate 8 a plurality of removal pipes 17 are inserted or are able to be inserted; however, for the sake of simplicity only one removal plate 17 is shown in the figure. For this purpose the pipe plate 8 preferably has pipe pieces 10 that are welded into it and can have the removal pipes 17 inserted into them. The removal pipes 17 inserted in this way are tightly welded with the pipe pieces 10. If defective removal pipes 17 must be replaced, these can be removed from the pipe plate 8 after removal of the weld seam. However, the welding described is not absolutely necessary, since under certain circumstances a fixed connection can be avoided altogether, or alternative connecting methods other than welding may be used.

[0021] In the second pipe plate 30, a number of reactor pipes 32 corresponding to the number of removal pipes 17 can be inserted or are insertable—once again only one reactor pipe 32 is shown in the figure for the sake of visibility. The reactor pipes 32 are also preferably inserted or insertable in the pipe pieces 31 welded into the pipe plate 30, wherein here also a great variety of connecting methods may be used.

[0022] The removal pipes 17 inserted into the reactor base 8 extend through the openings 11 and 20 into the reactor pipe 32 inserted in the second pipe plate 30.

[0023] Corresponding to an advantageous embodiment of the reactor in accordance with the invention, the reactor pipes 32 preferably extend through the base into the gas space 7 located beneath the second inner container 34.

[0024] The removal pipes 17 also advantageously have metal bellows 18 attached in the area of the openings 11, wherein these [bellows] are fastened with one of their ends to the removal pipes 17. Once again the fastening takes place preferably my means of welding, but here also alternative connection methods are conceivable. The removal pipes 17 are thus fixed slidingly in the area of the opening 11. The provision of a metal bellows 18 permits adequate assurance against large leaks between the first gas space 16 and the second gas space 21, since the metal bellows lie with their open ends on the base of the upper inner container 9. It is also conceivable that the open ends of the metal bellows 18 are connected with the base of the upper inner container 9 by means of a suitable mechanism.

[0025] The reactor pipes 32 are at least partially designed as membrane pipes 33. In this process the area of the reactor pipe 32 designed as membrane pipe 33 is preferably exclusively assigned to the second gas space 38.

[0026] The area of the reactor pipe 32 designed as a membrane pipe 33 can be formed either as a gas-tight, oxygen ion and electron-conducting ceramic membrane applied onto a gas-permeable support tube or produced in the form of a pipe made of a monolithic, gas-tight, oxygen ion and electron-conducting ceramic.

[0027] In the reactor design shown in the figure, the area of the reactor pipe 32 designed as a membrane pipe 33 is preferably connected coaxially and in a material-type fashion preferably at each of its ends with metal pipes of about the same diameter. The reactor pipes 32 are fixed only on one of their ends in the pipe plate 30, whereas the respective other end, to be sure closed gas-tight is guided or arranged freely in the expandable in the axial direction and able to slide so as to avoid stresses due to differences in thermal expansion. The same is true is for the removal pipes 17, which are likewise fastened only at one of their ends into the pipe plate 7 or the pipe pieces 10 arranged on these.

[0028] The lower inner container 34 in accordance with an advantageous designed of the reactor in accordance with the invention is at least partially filled with a preferably granular catalyst 35. In this process the lower inner container 34 is preferably filled to above the region of the reactor pipe 32 formed as the membrane pipe 33.

[0029] Alternatively or as a supplement to this, the reactor pipes 32, at least the areas of the reactor pipes 32 designed as a membrane pipe 33, can be coated with a catalyst.

[0030] The hot, oxygen-containing gas mixture is conveyed to the reactor in accordance with the invention over the opening 22 into the third gas space 21. The oxygen-containing gas mixture at a pressure of for example 1.5 bar has a temperature of 900°C. The production of such a gas mixture can for example take place in a combustion chamber under an access of fresh air. From the gas space 21 this gas mixture passes into the annular space 23 formed by the removal pipes 17 and reactor pipes 32.

[0031] The gas mixture flows along the area of the reactor pipes 32 designed as membrane pipes 33. In this process, pure oxygen passes through the membrane pipe 33 into the second gas space 38 and into the catalyst 35 arranged therein. To this second gas space 38, over the opening 36 a hydrocarbon mixture—if desired with addition of steam—with a temperature of about 500°C and a pressure of about 30 bar is introduced. The hydrocarbon mixture reacts by means of the catalyst 35 with the oxygen to form a syntheses gas, which leaves the second gas space 38 over the opening 37 and then the area 7 over the planned opening 6. The syntheses gas formed at a pressure of about 30 bar has a temperature of about 950°C.

[0032] Over the removal pipes 17, the area 4, and the opening 3 provided in the cover 2, a gas mixture reduced in oxygen is withdrawn from the reactor and possibly conveyed for further energy utilization. In this process the hot gas mixture reduced in oxygen is cooled in the removal pipes 17 in heat exchange with fresh air from about 950°C to about 160°C.

[0033] This cooling is achieved in that first through opening 25 fresh air that has a temperature of 90°C at a pressure of 1.7 bar is introduced into the fourth gas space 34, from which it is withdrawn via the opening 26 and then conveyed over the opening 12 to the first gas space 16. Since this gas space is in thermal contact with the removal pipes 17, the hot gas mixture reduced in oxygen is cooled in this way in the removal pipes 17. From the first gas space 16 the fresh air forcibly heated to a temperature of about 700°C is again withdrawn from the reactor over the opening 13 and, if desired, introduced to the previously mentioned combustion chamber for heating the oxygen-containing gas mixture.

[0034] Instead of fresh air, for the opening 25 steam may be introduced into the fourth gas space 24 for cooling and withdrawn over the opening 26. In this case the fresh air is introduced directly through the opening 12 into the first gas space 16.

[0035] The pipe plate 30, which is exposed to a pressure difference of 28.3 bar, is heated to a maximal temperature of 500°C in the reactor design in accordance with the invention.

[0036] In addition, in the reactor design in accordance with the invention, the higher pressure preferably prevails on the outside of the reactor pipe 32. This is advantageous, since in general the pressure resistance of ceramic is higher than its tensile strength.

[0037] Not shown in the figure are so called turbulence enhancers, which serve to improve material transfer and are preferably designed in the form of vortex-generating (guide) baffles. These turbulence enhancers can be arranged in the first gas space 16, the second gas space 38, within the removal pipes 17, preferably in the area of the first gas space 16, and/or in the annular gaps 23 formed by the removal pipes 17 and the reactor pipes 32.

[0038] The first alternative of the reactor in accordance with the invention differs from the second alternative of the reactor in accordance with the invention, explained on the basis of the figure, that the inner container 9—and thus also the gas space 16 that this defines—as well openings 12 and 13 are not present.

[0039] With regard to the loading of the membrane pipes 33 it is advantageous that the pressure of the syntheses gas and of the oxygen-containing gas mixture differ greatly. In the case of a low pressure difference, the gas mixture, reduced in oxygen, possibly leaving the reactor over the opening 3 must or should not be cooled, since this gas mixture—under relatively high pressure—can be conveyed for example to an expansion turbine for better energy utilization, wherein the highest possible gas temperature is reasonable.

[0040] The design in accordance with the first alternative therefore avoids the possibility of integrated cooling of the gas mixture reduced in oxygen.

[0041] The reactor constructions in accordance with the invention permit the achievement of adequate cooling preferably of all pressure-loaded metal components. Furthermore, a safe, gas-tight passage from the area of a reactor pipe designed as membrane pipe to the metal components of the reactor pipe is guaranteed. Furthermore, damaged reactor pipes can be replaced relatively easily and rapidly. Above and beyond this the reactors in accordance with the invention are constructed as conventionally as possible, so that the number of the ceramic components can be kept small. Furthermore, an improved reaction guidance is made possible, since the feed of the oxygen to the hydrocarbon or hydrocarbon mixture can take place over the pipe length, i.e., along the reaction route.

[0042] In addition to the membrane types mentioned, the reactors in accordance with the invention are also suitable for the use of other membranes, which can be integrated into the reactor designs in the manner described.

Claims

1. Reactor with

-a reactor jacket,

-two covers closing the two ends of the reactor jacket

-wherein each cover has at least one opening

-a first pipe plate arranged in the upper area of the reactor and a second pipe plate arranged in the center area of the reactor

-an inner container that is arranged beneath the second pipe plate and defines a gas space

-wherein the inner container may have openings in its base

-at least one partition arranged essentially at a right angle to the reactor jacket, having openings, which subdivides the area between the first pipe plate and the second pipe plate into a gas space arranged above the partition and a gas space arranged below the partition

-in each case at least two openings corresponding to the gas space arranged below the partition and to the gas space defined by the inner container, wherein in the case of the gas space arranged below the partition, these are preferably arranged in the reactor jacket, and in the case of the gas space defined by the inner container, preferably an opening is arranged in the reactor jacket and openings in the base of the inner container

-at least one opening corresponding to the gas space arranged the above the partition

-several removal pipes placed in the first pipe plate

-several reactor pipes placed in the second pipe plate

-wherein the removal pipes extend through the openings into the reactor pipes

-wherein the reactor pipes are at least partially designed as membrane pipes, and

-wherein the region of the reactor pipes designed as membrane pipes is arranged in the gas space defined by the inner container.

2. Reactor with

a reactor jacket (1, 1')

-two covers (2, 5)closing off the two ends of the reactor jacket (1,1'),

-wherein each cover (2,5) has at least one opening (3, 6)

-a first pipe plate (8) arranged in the upper area of the reactor and a second pipe plate (30) arranged in a middle area of the reactor

-an upper and a lower inner container (9, 34) each of which is arranged beneath the pipe plates (8, 30) and defines a first and a second gas space (16, 38)

-wherein at least the upper inner container (9) has openings (11) in its plate

-at least one partition (19) arranged essentially at right angles to the reactor jacket (1,1') beneath the inner container (9), having openings (20), and subdividing the area between the upper inner container (9) and the second pipe plate (30) into a third (21) and a fourth (24) gas space

-in each case at least two openings (12, 13, 36, 37, 25, 26) corresponding to the first, second, and fourth gas space (16, 38, 24), wherein these are arranged preferably in the reactor jacket (1') in the case of the first and the fourth gas space (16, 24), and in the case of the second gas space (38), preferably one opening (36) is arranged in the reactor jacket (1)and openings (37) are arranged in the base of the lower inner container (34)

-at least one opening (22) corresponding to the third gas space (21)

-several removal pipes (17) inserted in the first pipe plate (8)

-several reactor pipes (32) arranged in the second pipe plate (30)

-wherein the removal pipes (17) extend through the openings (11) into the reactor pipes (32)

-wherein the reactor pipes (32) are at least partially designed as membrane pipes (33),

and

-wherein the area of the reactor pipes (32) designed as a membrane pipe (33) is arranged in the second gas space.

3. Reactor in accordance with Claim 1 or 2, characterized in that the area of the reactor pipe (32) formed as a membrane pipe (33) is designed in the form of a gas-tight, oxygen ion- and electron-conducting ceramic membrane applied onto a gas-permeable support pipe.
4. Reactor in accordance with Claim 1 or 2, characterized in that the area of the reactor pipe (32) designed as a membrane pipe (33) is produced in the form of a pipe consisting of a monolithic, gas-tight, oxygen ion- and electron-conducting ceramic.
5. Reactor in accordance with one of the preceding claims, characterized in that the lower inner container (34) is at least partially filled with a preferably granular catalyst (35).

6. Reactor in accordance with one of the preceding claims, characterized in that the reactor pipe (32), at least the area of the reactor pipe (32) formed as a membrane pipe (33), is coated with a catalyst.
7. Reactor in accordance with one of the preceding claims, characterized in that the reactor pipes (32) extend through the base of the lower inner container (34) into the lower gas space (7).
8. Reactor in accordance with one of the preceding claims, characterized in that on the inside of the reactor jacket (1, 1') and/or on the inside of one cover or the covers (2,5) a thermal insulating layer is arranged.
9. Reactor in accordance with one of the preceding claims, characterized in that on the pipe plate or plates (8, 30), pipe pieces (10, 31) are arranged, into which the removal pipes (17) and/or the reactor pipes (32) can be inserted.
10. Reactor in accordance with one of the preceding claims 2-9, characterized in that in the area of the openings (11) provided in the base of the upper, inner container (9) the removal pipes (17) have gas-tight attached metal bellows (18), wherein these are fastened with one of their ends on the removal pipes (17), preferably gas-tight.
11. Reactor in accordance with Claim 10, characterized in that the metal bellows (18) are welded with the removal pipes (17).
12. Reactor in accordance with one of the preceding claims, characterized in that in the first gas space (16), the second gas space (38), inside the removal pipes (17), preferably within the removal pipes (17) in the area of the first gas space (16) and/or in the annular gaps (23) formed by the removal pipes (17) and the reactor pipes (32), turbulence enhancers are arranged.
13. Reactor in accordance with one of the preceding claims, characterized in that the reactor is formed in a cylindrically symmetrical manner.

One page of drawings attached.

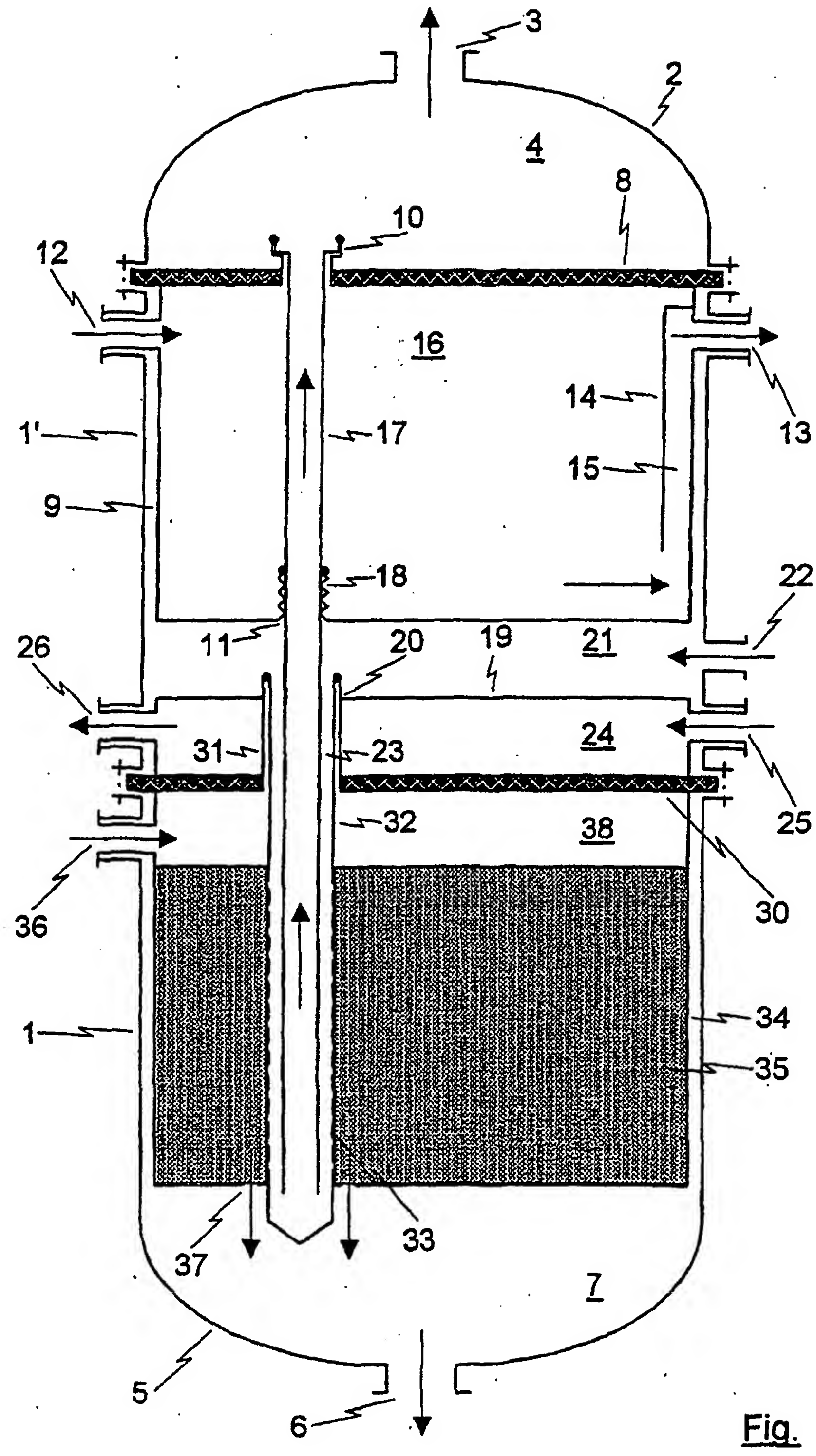
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Fig.